Spatial assessment of heavy metals contamination in household garden soils in rural Limpopo Province, South Africa

Thandi Kapwata[a,b*], Angela Mathee[a,b,c], Neville Sweijd[d], Noboru Minakawa[e], Mirriam Mogotsi[a], Zamantimande Kunene[a] and Caradee Y Wright[f,g]

a. Environment and Health Research Unit, South African Medical Research Council, Johannesburg 2028, South Africa
b. Environmental Health Department, Faculty of Health Sciences, University of Johannesburg, Johannesburg 2028, South Africa
c. School of Public Health, University of the Witwatersrand, Johannesburg 2028, South Africa
d. Applied Centre for Climate and Earth Systems Science, National Research Foundation, Cape Town, South Africa. nsweijd@access.ac.za
e. Institute of Tropical Medicine, Nagasaki University, Nagasaki, Japan
f. Environment and Health Research Unit, South African Medical Research Council, Pretoria 0084, South Africa. cwright@mrc.ac.za
g. Department of Geography, Geoinformatics and Meteorology, Private Bag X20, Hatfield, Pretoria 0028, South Africa

* Corresponding author: Thandi.Kapwata@mrc.ac.za

Abstract

Heavy metal pollution in soil poses a serious health threat to humans living in close proximity and in contact with contaminated soil. Exposure to heavy metals can result in a range of adverse health effects, including skin lesions, cardiovascular effects, lowering of IQ scores and cancers. The main objectives of this study were to i) use a portable XRF spectrophotometer to measure concentrations of lead (Pb), arsenic (As), mercury (Hg) and cadmium (Cd) in residential soils in rural Giyani in the Limpopo province of South Africa; ii) to assess the spatial distribution of soil metal concentrations; iii) to assess pollution levels in residential soils. There were elevated levels of As at one of the sites where 54% of soil samples exceeded the Canadian reference levels for As of 20 mg/kg. Using the geoaccumulation index (I_{geo}) to determine contamination levels of As, 57% of soil samples from the most polluted site were found to be moderately to heavily and extremely contaminated with As (I_{geo} class 2 to 5). The site is located near the Giyani Greenstone Belt, which is characterized by abandoned mines and artisanal mining activities. Gold ores are

closely associated with sulphide minerals such as arsenopyrite and these have been found to contain high amounts of As. This study highlighted the potential for soil contamination and the importance of site-specific risk assessment in the context of environment and health impact assessments prior to major developments, including human settlement developments.

Keywords: metal pollution, contaminated residential soil, arsenic, health risk.

1. Introduction

Heavy metal pollution in soil is a serious environmental health concern due to potential toxicity at trace level, bioaccumulation, persistence and the lack of biodegradability of heavy metals (Grandjean and Landrigan, 2014, Facchinelli et al., 2001, Li et al., 2006, Jia et al., 2018). Exposure to elevated levels of heavy metals have been linked to various adverse human health effects. Studies found that lead and arsenic were associated with neurological impairment among children in the United States of America (Canfield et al., 2003), Bangladesh (Wasserman et al., 2004) and Mexico (Rosado et al., 2007). Chronic exposure to nickel (Ni), cadmium (Cd) and arsenic (As) may cause lung cancer (Zhao et al., 2012). Exposure to elevated levels of heavy metals may also result in kidney dysfunction, hypertension, respiratory illnesses, increased risk of foetal death and impaired growth, bone fractures and skin lesions (Zhao et al., 2012, Llanos and Ronco, 2009).

Heavy metals have long residence and persistence times in soil, thus increasing the potential or duration of risks for human exposure and harmful effects (Fetter et al., 2017). Metal contamination of soils may be caused by natural and anthropogenic sources. Natural processes that release heavy metals into soil include weathering of rocks, volcanic eruptions and atmospheric depositions (Hooda, 2010). Human activities that affect the distribution and content of heavy metals in soils include formal and informal or artisanal mining, industrial processes, use of fossil fuels including vehicle exhausts, the operation of cottage industries, agriculture (use of pesticides, fertilizers, sewage sludge and livestock manures) and waste disposal (Zhang et al., 2009, Li et al., 2006, Nicholson et al., 2003, Atafar et al., 2010, Teare et al., 2015). Studies show that elevated concentrations of heavy metals in soil pose a risk to human health through various exposure routes: direct ingestion of soil, inhalation of dust, consumption of plants grown in contaminated soil and dermal contact (Zhang et al., 2009, Liu et al., 2013, Farmer et al., 2011, Zheng et al., 2010, Siciliano et al., 2009, Luo et al., 2012).

In recent years, portable X-ray fluorescence (XRF) spectrometers have been used extensively to monitor soil contamination by heavy metals (Ackah, 2019, Caporale et al., 2018, Meza-Montenegro et al., 2012, Rinklebe et al., 2019, Akopyan et al., 2018). XRF spectroscopy has been found to provide accurate total elemental contents in solid soil samples (Reidinger et al., 2012). Other advantages of using portable XRF spectrometers to measure soil metal concentrations instead of conventional laboratory based chemical analyses include cost effectiveness, results are available within a short amount of time and ease of analysis (Caporale et al., 2018, Chakraborty et al., 2017).

Previous studies in South Africa have assessed soil metal concentrations in urban areas. Mathee et al. (2018) reported levels of lead (Pb) and As exceeding local and international guidelines in garden soils in residential areas in Johannesburg's inner city as well as close to a mine tailings facility. High levels of As were also found in soils collected from a school garden in Johannesburg (Kootbodien et al. (2012)), while metal concentrations in soils in the Tshwane area of Pretoria showed high levels of trace metals in samples collected close to high traffic density and industrial areas (Olowoyo et al. (2010)). Other studies have focused on the concentrations of metals including Mn, Cu, Cd, Co, Zn, Ni, As, Pb, Hg and Cr in soil around mine tailings in South Africa (Mitileni et al., 2011, Rembuluwani et al., 2014, Kamunda et al., 2016, Ngole-Jeme and Fantke, 2017, Lusilao-Makiese et al., 2013) However, there are no studies that have assessed the spatial distribution of heavy metals in soils from rural residential areas of Limpopo province, South Africa. The main objectives of this study were to i) use a portable XRF spectrophotometer to measure concentrations of Pb, As, Hg and Cd in residential soils in rural Giyani in the Limpopo province of South Africa; ii) to assess the spatial distribution of soil metal concentrations; iii) to assess pollution levels in residential soils.

2. Materials and methods

2.1. Study location and soil sampling

A total of 310 soil samples were collected during February 2017 in four sites in Giyani, a town located in the Greater Giyani Municipality of Limpopo Province, South Africa. The study sites were Dzingidzingi (site 1), Maswanganyi (site 2), Siandana (site 3) and Tomu (site 4), comprising a total population of 14 500 people and 3784 dwellings (Stats SA, 2011). Random sampling was used to select dwellings within each site. Composite soil samples were obtained from the top 10 cm of exposed residential garden soils in accordance with United States (US) Environmental Protection Agency (EPA) operating procedure (USEPA, 2014). After collection, soil samples were oven-dried at 40 ˚C over 48 hours and sieved to particles less than 2 mm. The concentrations of Pb, As, Hg and Cd were measured using a portable X-ray fluorescence (XRF) spectrometer (Hu et al., 2014). The limits of detection were 7 mg/kg for Pb and Hg; 5 mg/kg for As and 10 mg/kg for Cd (Niton XRF, 2015).

2.2. Statistical Analysis

The mean, range, standard deviation (SD) and median concentrations of Pb, As, Hg and Cd were calculated for each site. The percentage of soil samples exceeding local and international guidelines were also calculated. A one-way ANOVA was conducted to determine if concentrations of Pb, As and Hg were statistically different across the study sites (Cd was excluded because measured concentrations were all below the limit of detection). All descriptive statistics and ANOVA calculated using STATA version 14.0 (StataCorp, 2017).

2.3. Index of geo-accumulation

The geoaccumulation index (I_{geo}) is a single index that uses only one metal contaminant to give an indication of the pollution level of that metal. This index has been widely used to assess the levels of heavy metal contamination (Jia et al., 2018, Zhu et al., 2017). I_{geo} was first proposed by Muller (1969) and is calculated using the following equation:

$$I_{geo} = \log_2 \frac{C_n}{1.5\ X\ B_n} \qquad \textit{(Equation 1)}$$

where C_n is the average concentration of metal in the soil and B_n is the background concentration of the metal. The factor 1.5 was introduced to minimize the effect of possible variations in baseline data which might be attributed to lithologic variations in the soils (Solgi et al., 2012). The I_{geo} index consists of seven classes from 0 (uncontaminated) to 6 (extremely contaminated) and these are listed in Table 1.

Table 1: Seven classes comprising the geoaccumulation index as classified by Muller (1969)

I_{geo} Value	**I_{geo} Class**	**Level of contamination**
< 0	0	Uncontaminated
0 – 1	1	Uncontaminated to moderately contaminated
1 - 2	2	Moderately contaminated
2 - 3	3	Moderately to heavily contaminated
3 - 4	4	Heavily contaminated
4 - 5	5	Heavily to extremely contaminated
> 5	6	Extremely contaminated

2.4. GIS

ArcGIS® software (ESRI, 2010) was used to assess and display the spatial distribution of heavy metals.

2.5. Ethical considerations

Research ethics clearance for the study was granted by the South African Medical Research Council Ethics Committee (Certificate number: EC005-3/2014, re-approval 26 June 2018).

3. Results

The World Health Organization (WHO) lists lead, arsenic, mercury and cadmium as four of the 10 chemicals of major public concern (WHO, 2010). Descriptive statistics for Pb, As, Hg and Cd concentrations of garden soils in rural areas in Limpopo province are listed in Table 2. The South African and Canadian screening and reference levels of metals in soil are also given in the table (DEA, 2010, CMoE, 2011). The maximum concentrations of all heavy metals at sites 3 and 4 were below the recommended values. The maximum concentration of Hg for site 1 was above the screening level for only one sample out of 96. Concentrations of Hg and other metals in the remaining 95 collected samples were below screening levels. There were elevated levels of As at site 2, with 10% of collected samples exceeding the South African screening level of 48 mg/kg, while 23% of samples had As concentrations exceeding 40 mg/kg. However, 54 % of soil samples exceeded the Canadian reference level for As of 20 mg/kg (CMoE, 2011). The ANOVA results showed that there was a statistically significant difference in As concentration across the 4 sites ($p < 0.001$). However, the differences in Pb and Hg concentrations were not statistically significant ($p = 0.50$ for both).

Table 2: Summary of heavy metals content (all concentrations reported in mg/kg)

Site	Metal	Mean	Range	Median	South African Soil Screening Values#	Canadian reference level$	% of samples exceeding South African guideline level	% of samples exceeding Canadian reference level
Site 1 N = 96	Pb	12.2	0 – 113.3	15.9	230	120	0	0
	As	0.2	0 – 17.2	17.2	48	18	0	0
	Hg	0	0 – 11.0	11.0	1	1	1.0	1.0
	Cd	<LOD*	-	-	32	1.2	0	0
Site 2 N = 95	Pb	5.1	0 – 111.7	19.4	230	120	0	1.1
	As	24.5	0 – 173.0	36.8	48	18	9.5	53.7
	Hg	17.8	16.5 – 19.0	17.8	1	1	2.1	2.1
	Cd	<LOD*	-	-	32	1.2	-	-
Site 3 N = 98	Pb	14.7	0 – 144	16.0	230	120	0	1.0
	As	<LOD*	-	-	48	18	-	-
	Hg	<LOD*	-	-	1	1	-	-
	Cd	<LOD	-	-	32	1.2	-	-
Site 4 N = 21	Pb	12.8	0 – 23.	18.3	230	120	0	0
	As	<LOD*	-	-	48	18	-	-
	Hg	<LOD	-	-	1	1	-	-
	Cd	<LOD*	-	-	32	1.2	-	-

Note. * LOD: limit of detection. # The soil screening levels are to provide norms and standards for the identification and registration of contaminated sites and are not an indication of what is allowed in terms of human health. $ Reference levels are for general guidance only for the protection of environmental and human health

The concentrations of heavy metals in soils analysed from sites 1, 3 and 4 were all below the South African soil screening levels (Table 2), except for a high Hg reading for a single sample (1/96) from site 1. However, elevated As concentrations in soils sampled from site 2 (Maswananyi) were more widespread, therefore the geo-accumulation index was calculated only for As at site 2 (Table 3). Results showed that 57 % of collected soil samples were moderately to extremely contaminated with As (I_{geo} class 2 to 5).

Table 3: Levels of contamination in garden soil samples due to Arsenic

Igeo_class	Level of contamination	Number of soil samples n*	Percentage %
0	Uncontaminated	20	39
1	Uncontaminated to moderately contaminated	2	4
2	Moderately contaminated	10	20
3	Moderately to heavily contaminated	15	29
4	Heavily contaminated	3	6
5	Heavily to extremely contaminated	1	2
6	Extremely contaminated	0	0

*Total n=51 because I_{geo} was not calculated for samples that contained As that were below limit of detection (7mg/kg).

Figure 1 presents the spatial distribution of the I_{geo} index values and concentrations of As at site 2, the site with elevated levels of As. A north-south distribution gradient is evident in the spatial distribution of As with higher concentrations being observed in the north and lower concentrations in the south.

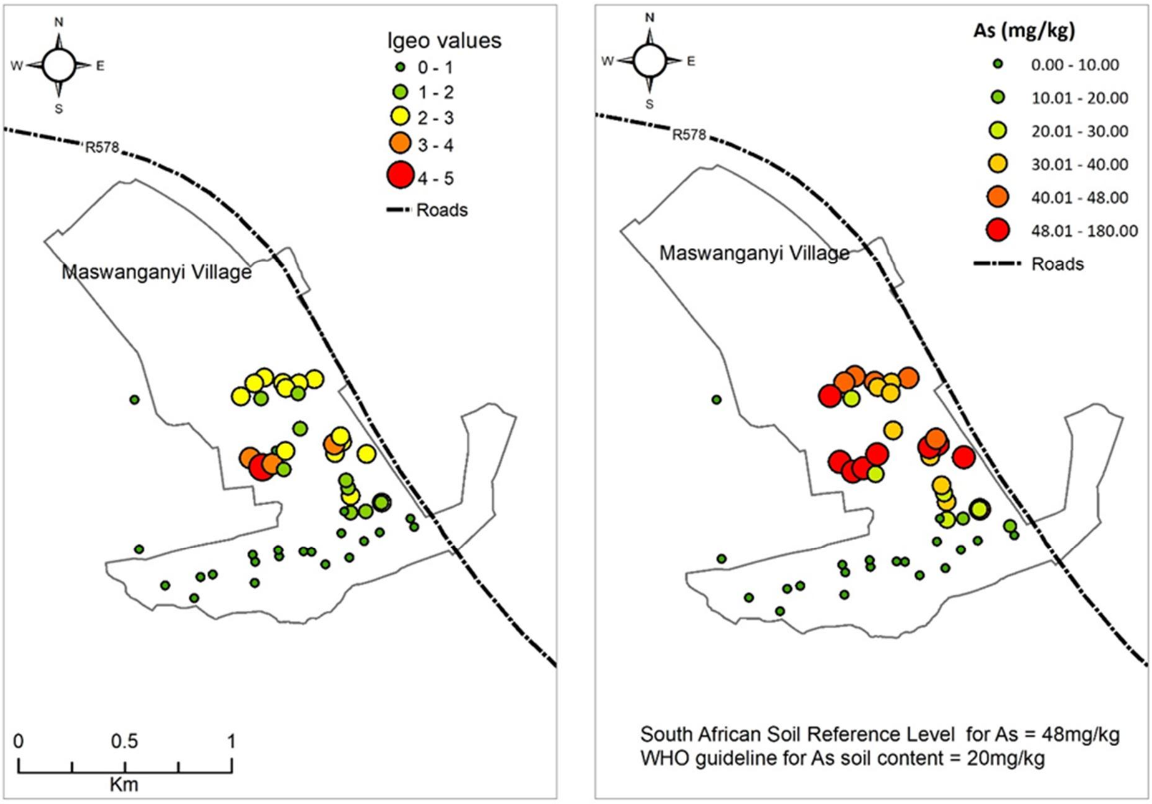

Figure 1: Spatial distribution of the Igeo index values and concentrations of As at site 2 – Maswanganyi

Further investigation and interrogation of the map in Figure 2 showed that samples with high As levels at site 2 are found on land identified as a possible gold mining area located in close proximity to the Giyani Greenstone Belt (GGB) that is comprised of several abandoned mines (Steenkamp and Clark-Mostert, 2012). In addition to this, there is anecdotal evidence of illegal and artisanal mining in decommissioned mines in the area.

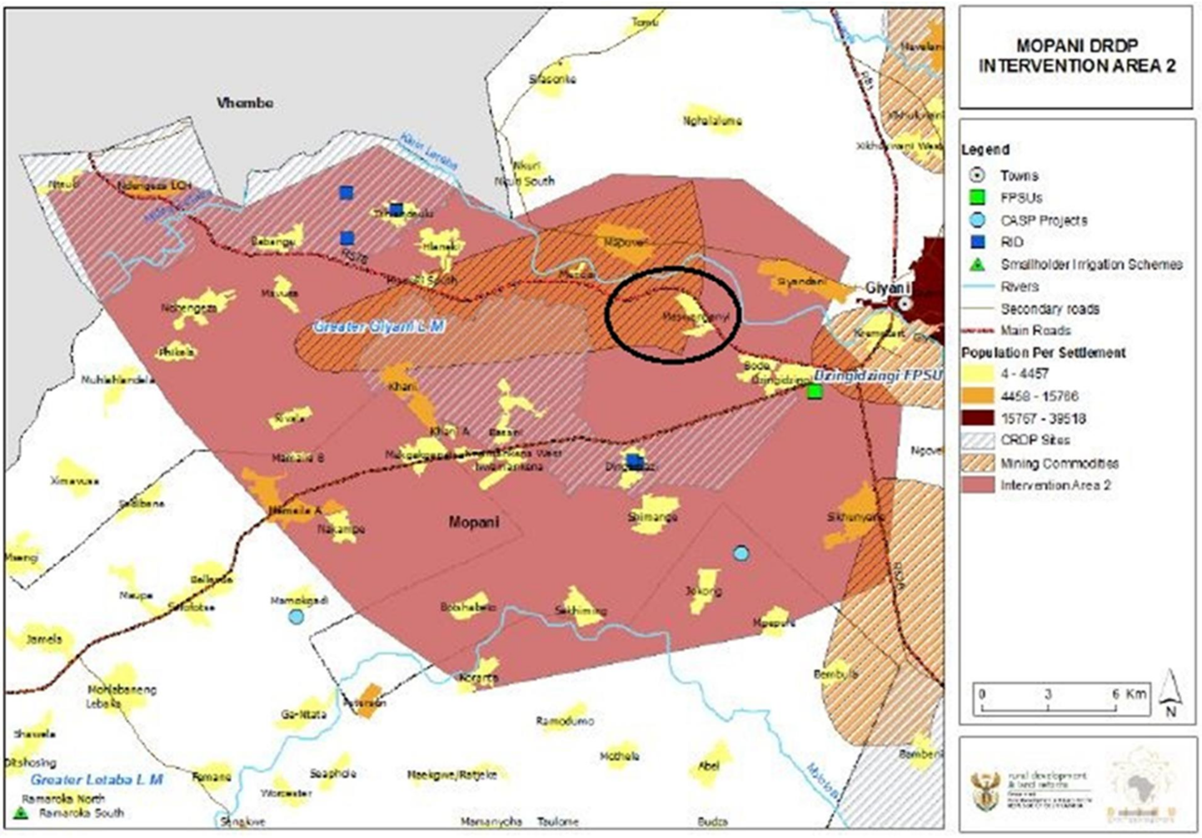

Figure 2: Mining commodities (land identified to have mineral resources) in Mopani, Limpopo province (Source: Department of Rural development and land reform), study site depicted in black circle.

4. Discussion

The majority of concentration levels of Pb, As, Hg and Cd at sites 1,3 and 4 were below the South African screening levels. However, relative to the remaining three sites, a significantly higher proportion of soil samples with elevated levels of As were found in Maswanganyi (site 2), where As levels in 9% of samples exceeded the South African screening levels, and 54 % exceeded the Canadian reference levels (CMoE, 2011). The geo-accumulation index (I_{geo}) was used to assess the degree of contamination of the soil; it showed that 57% of soil samples collected in Maswanganyi were contaminated by As.

The town of Giyani is in the Giyani Greenstone Belt which consists of several abandoned, existing and prospective gold mines (Steenkamp and Clark-Mostert, 2012, Carranza et al., 2015), and where artisanal mining has anecdotally been reported to be occurring. Spatial analysis of the distribution of As identified a pattern of higher concentrations of As in the samples collected in the north of Maswanganyi and lower concentrations to the south. The Greater Giyani Municipality is well known for gold mineralization (Steenkamp and Clark-Mostert, 2012, Carranza et al., 2015) and gold deposits are reported to have been identified in the northern part of Maswanganyi (Figure 2). Therefore a possible source of the elevated levels of As could be the local gold-bearing ores (Maseki et al., 2017).

Gold ores are closely associated with sulphide minerals such as arsenopyrite and these have been found to contain high amounts of As (García-Lorenzo et al., 2012). Previous studies have found high As content in soils around gold mines. A study evaluating As concentrations in soils near artisanal and small scale gold mines in Bolivia found that mean concentrations ranged from 13 to 64 mg kg^{-1}(Acosta et al., 2015). This was above 12 mg kg^{-1} which is the maximum allowed level recommended by the Canadian Soil Quality Guidelines (CCME, 1999). Soil samples collected around a gold mining district in Iran found high concentrations of As with a mean concentration of 54.12 mg kg^{-1}, the associated geoaccumulation index of 4.33 showed that the study area was heavily contaminated with As (Keshavarzi et al., 2012). Similar findings were reported in Australia where the median As concentration of soil collected from a legacy mining township was 85 mg kg^{-1} which was 28 times higher than the Australian top soil average of 3 mg kg^{-1}(Abraham et al., 2018).

Soil samples from mine tailings and sediment in Malamulele, which is in close proximity to the study site (Giyani), have been shown to contain elevated levels of heavy metals including Cu, Co, Cd, As, Zn, Ni and Mn (Rembuluwani et al., 2014, Mulugisi et al., 2009). Another study assessing metal content of soil collected around a mine tailings dam also in Malamulele found high As levels, with concentrations five to six times above background soil levels (Bowen, 1979, Muzerengi, 2017). Wind and water erosion of mine tailings and rock flour enriched with arsenopyrite minerals could be responsible for the As enrichment in soils resulting in the contamination at site 2 (Matshusa et al., 2012, Acosta et al., 2015). It is also possible that contamination of soil in Maswanganyi could have occurred through the transportation of soil particles and metals from nearby mine tailings facilities.

The soil environment has been identified as a contributor to arsenic human exposure (Pearce et al., 2010, Hinwood et al., 2003). Ingestion, especially through hand-mouth contact in children, is the main route of exposure (Li et al., 2001). Chronic exposure to As has been associated with significant health consequences including dermatological effects, cardiovascular effects, respiratory disorders, pulmonary disorders, neurological effects and cancer (Pearce et al., 2012, Abdul et al., 2015). The effects of As are divided into four stages - preclinical (characterized by high levels of As in biological samples with no clinical symptoms); clinical (arsenic toxicity confirmed after biological samples are tested, visible clinical symptoms begin to develop); internal complications (functioning of internal organs begins to be affected) and malignancy (development of tumours and other complications that may eventually result in death) (Abdul et al., 2015). Clinical and epidemiological studies have found that long-term exposure to As is also associated with increased risks for diabetes, high blood pressure, respiratory diseases (Parvez et al., 2011), cardiovascular diseases (Chen et al., 2011) and neurological effects such as a feeling of "pins and needles" (paresthesia) in the hands, impairment of memory, hallucinations, seizures and even Alzheimer's disease (Vahidnia et al., 2007, O'Bryant et al., 2011), as well as lower IQ in children (CDC, 2017). Arsenic exposure has also been found to be associated with skin disorders, such as darkening (hyperpigmentation) or thickening of the skin (hyperkeratosis), skin corns (type of callus), warts (growth of the skin) and dermal lesions (CDC, 2007, Chakraborti et al., 2013, Żukowska and Biziuk, 2008). Ingestion of elevated levels of inorganic arsenic may also cause gastro-intestinal symptoms such as nausea, vomiting and diarrhoea. Arsenic is also a known carcinogen that has been associated with several cancers including skin (Yu et al., 2000, Surdu, 2014), liver (Wang et al., 2014, Liaw et al., 2008), bladder (Steinmaus et al., 2014), kidney (Ferreccio et al., 2013), lung (Hubaux et al., 2013, Lamm et al., 2013) and prostate cancers (Siegel et al., 2013). Furthermore, recent studies have provided evidence of As-induced epigenetic modifications that can have adverse effects on human health (Bjørklund et al., 2018, Bailey et al., 2013).

The levels of arsenic identified in residential soil samples collected from Maswanganyi in this study indicate the potential for significant public health risks and underscores the importance of a precautionary approach to the location of human settlements in relation to local sources of pollution. In this regard, informed decision-making, for example using environment and health impacts assessments, as well as inter-sectoral planning and action, in urban as well as rural areas in South Africa and elsewhere, are likely to afford communities higher levels of environmental health protection than is currently the case.

5. Conclusions

The concentrations of four heavy metals (Hg, As, Cd and Pb) in residential soils sampled from 310 locations across four sites were measured using a portable X-ray fluorescence spectrometer (XRF). The results showed that there were low concentrations of all metals in three sites therefore no or minimal heavy metal contamination can be expected in those sites. However, one site had high concentrations of As which exceeded international recommendations therefore the geoaccumulation index was used to determine the levels of As pollution for that site. According to this index, the soil showed moderate to high contamination of As. High levels of heavy metals in residential soil is a serious public health risk because heavy metals that have been substantially accumulated in soils can release to other ecosystems, such as groundwater, rivers, atmosphere and crops, and consequently are hazardous to human health and the environment. Detection of elevated levels of heavy metals in soil in existing residential areas is necessary to identify high risk communities. Failure to control population exposure to heavy metals in contaminated soil is likely to significantly impact public health. This study highlighted the importance of site-specific risk assessment due to the wide variation of heavy metal concentrations across the four sites near a single town. Soil contamination assessments should be conducted prior to human settlement development, and thereafter only regularly tested if there is reason to believe that the situation may have changed.

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Author contributions. AM and CYW conceptualised the study; ZK and MM performed the data collection and sample testing; TK performed the data analysis and presentation; all authors contributed to the writing of the manuscript.

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